

# Nonlinear acoustic and microwave absorption in disordered semiconductors

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Nonlinear hopping absorption of ultrasound and electromagnetic waves in amorphous and doped semiconductors is considered. It is shown that even at low amplitudes of the electric (or acoustic) field the nonlinear corrections to the relaxational absorption appear anomalously large. The physical reason for such behavior is that the nonlinear contribution is dominated by a small group of close impurity pairs having one electron per pair. Since the group is small, it is strongly influenced by the field. An external magnetic field strongly influences the absorption by changing the overlap between the pair components' wave functions. It is important that the influence is substantially different for the linear and nonlinear contributions. This property provides an additional tool to extract nonlinear effects.

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## I. INTRODUCTION

The subject of this article is nonlinear microwave and acoustic properties of amorphous semiconductors and lightly doped crystalline semiconductors in the regime of hopping conductance. We are interested in absorption to electron transitions between localized states associated with defects or impurity atoms. We consider the case where the absorption is due to electron hopping within pairs of *neighboring* defects containing one electron per pair. The distance between the centers within the pair must be small enough to allow tunneling, while the distance to other impurities should be large enough to prevent tunneling to impurities outside the pair. In a weakly doped semiconductor we can expect these pairs to be relatively rare, and triplets of the same kind will be even less likely. Thus a natural basis to treat the problem is the so-called two level approximation according to which only the lowest energy level of each of two neighboring impurities are taken in consideration. This approach, as well as its range of applicability, was first discussed in detail by Pollack and Geballe.<sup>1</sup>

For brevity, in the following we shall discuss the case of acoustic attenuation and then specify what changes in the formulae should be introduced to allow for electromagnetic absorption.

An external AC electric or acoustic field causes transitions between the electron states. Direct inter-level transitions leading to absorption of quanta give rise to the so-called *resonant* absorption. For low intensities, the resonant contribution to the absorption coefficient of an acoustic wave can be expressed as<sup>2</sup>

$$\Gamma^{(\text{res})} = \alpha_1 \omega / s \tanh(\hbar\omega/2k_B T) \quad (1)$$

where  $\alpha_1$  is a dimensionless coupling parameter, weakly dependent on temperature and frequency,  $s$  is the sound velocity,  $\omega$  is the sound frequency, and  $T$  is the temperature. We can always assume the relation  $\hbar\omega/2k_B T \ll 1$ .

$\alpha_1$  will be specified later, see Eq. (18). Defining  $n_0$  as the Fermi function  $n_0(E) = [\exp(E/k_B T) + 1]^{-1}$ , we can write the factor  $\tanh(\hbar\omega/2k_B T)$  as  $n_0(\hbar\omega/2) - n_0(-\hbar\omega/2)$ . This can be recognized as the difference in the equilibrium population of the two levels of the pair with an energy splitting of  $\hbar\omega/2 - (-\hbar\omega/2) = \hbar\omega$ , that is, the pairs which can directly absorb a phonon.

The *relaxation* absorption is due to a modulation of the electron inter-level spacing  $2\epsilon$  by the AC field. Such a modulation leads to a periodic change of the occupation numbers of the two levels which lags in phase the variation of  $\epsilon$ . This lag leads to the energy dissipation. In the linear regime the coefficient of relaxation absorption has been calculated as<sup>3</sup>

$$\Gamma^{(\text{rel})} \approx \frac{\alpha_2}{s} \begin{cases} \tau^{-1}, & \omega\tau_0 \gg 1 \\ \omega, & \omega\tau_0 \ll 1 \end{cases} \quad (2)$$

where  $\alpha_2 \approx \alpha_1$  and  $\tau_0$  represents a minimal relaxation time for  $\epsilon \approx k_B T$ . The physical meaning of  $\tau_0$ , as well as estimates of this quantity, will be discussed later.

Comparing Eq. (2) with Eq. (1) we conclude that the relaxation absorption always predominates at  $\omega\tau_0 \ll 1$ . If  $\omega\tau_0 \gg 1$  the ratio  $\Gamma^{(\text{res})}/\Gamma^{(\text{rel})} \approx \omega\tau_0 \tanh(\hbar\omega/2k_B T)$  can be either greater or less than one under experimentally accessible conditions.

For higher intensities this comparison is no longer valid, as both the resonant and the relaxation absorption show strongly nonlinear behavior. For the resonant absorption, the nonlinearity is due to an equalization of the population numbers of the two electron states. From Eq. (1) we see that this leads to a strong reduction of the resonant absorption. Usually this suppression takes place at very low intensities.<sup>3</sup>

The nonlinearity of the relaxation absorption is due to the following. In the limit of high intensities, when the perturbing potential is amplitude  $\gtrsim k_B T$ , there will be times when  $2\epsilon(t) \leq k_B T$  and both states are almost equally occupied. During such part of the wave period no

transitions and thereby no absorption will occur. Consequently, the absorption coefficient decreases with the sound amplitude.

The linear relaxation absorption in semiconductors has previously been studied both theoretically<sup>4</sup> and experimentally (for a review, see Ref. 5), while for the strongly nonlinear regime only theoretical results<sup>2</sup> were obtained. The reason why this type of nonlinearity has not been observed experimentally is probably some masking by other mechanisms leading to a nonlinear behavior. One of such mechanisms could be wave-induced ionization of impurity atoms into the conduction band observed in InSb.<sup>6</sup> Consequently, to observe the mechanism<sup>2</sup> of nonlinear absorption one should carefully choose the material. That does not seem to be an easy task, and we are not aware of any experiments of this type.

In this article we will address the case of *low* intensities when nonlinear effect manifest themselves as small corrections to the linear absorption. The main message is that already the lowest order corrections offer interesting information and anomalous effects worth studying. Hopefully, these effects will be pronounced within an experimentally achievable parameter space where other mechanisms of nonlinear behavior are still not important.

The two-site approximation for semiconductors allows us to describe our system in terms of the so-called Two Level System (TLS) model. This model was first proposed independently by Anderson *et al.*<sup>7</sup> and Philips<sup>8</sup> to explain the low temperature specific heat in amorphous materials, and has been successful in describing several other phenomena. The model has given a theoretical explanation for a surprising universality in the behavior of very different disordered materials at low temperatures.

The authors<sup>9</sup> have previously used the TLS model to analyze nonlinear corrections to the absorption in both dielectric and metallic glasses. However, there are several important differences between the situations in glasses and in disordered or doped semiconductors. The main difference is due to the electric charge of the particle involved in the transitions. This introduces two modifications to the earlier results.

First, the electric charge gives the pair a dipole moment proportional to the distance between the pairs, the coupling being proportional to the dipole moment's component along the direction of the electric field. This leads to a specific orientational dependence of the absorption of one pair, and after integration over all pairs it significantly influences the absorption. Second, we must expect our system to behave differently with the application of a magnetic field. More specifically, as the magnetic field leads to a stronger localization of the electrons, we must expect the absorption to decrease with an applied field. The effect of a magnetic field will therefore also be analyzed, in the special cases of a weak or a strong field. The connection between the orientation of the dipole moment and the magnetic field direction also leads to a dependence of the absorption coefficient on the relative directions of the radiation wave vector and the magnetic field.

We will solve the problem for a magnetic field parallel or perpendicular to the wave vector of the radiation.

The magnetic field dependence gives us the possibility to separate the relaxational absorption from other contributions. It will also be shown that the effect of a magnetic field depends on the specific relaxation mechanism, thus providing us with a tool to further understand the relaxation processes for localized carriers.

The paper is organized as follows. First, we give a short introduction to the theory based on the TLS model and how it can be used to solve the problem of relaxation absorption in glasses. Then we will fit the two-site approximation for a semiconductor to the TLS model and show what modifications are needed for this. Finally, we will show how the effects of a magnetic field can readily be included in the analysis, and what effects to expect from this.

## II. TWO LEVEL SYSTEMS AND RELAXATION LOSSES IN GLASSES

The TLS model deals with a particle (in our case, an electron) moving in a slightly asymmetric double well potential. It is assumed that only the two ground levels are accessible. The ground levels in the isolated wells are assumed to have a slight separation in energy,  $2\Delta$ , and to be coupled via a tunneling energy overlap integral  $\Lambda$ . The Hamiltonian of such a system is traditionally written as

$$\mathcal{H}_0 = \Delta \sigma_z - \Lambda \sigma_x \quad (3)$$

where  $\sigma_i$  are the Pauli matrices.

Let us now apply an external periodic perturbing potential and study the power absorbed by a single pair,  $p(\Delta, \Lambda)$ . As it will be shown, this power is in general a non-monotonous function of its parameters, and there exists an “optimal” region which dominates absorption. What is important is that those regions are *different* for the linear absorption and for the nonlinear correction. Let us define  $\delta$  as the typical value of the inter-level splitting  $\Delta$  which is important for the onset of nonlinear behavior. Estimates for the quantity  $\delta$  will be given in the discussion section.

At  $\hbar\omega \ll \delta$  one can employ the adiabatic approximation and neglect time derivatives of the external field while solving the Schrödinger equation for the TLS.<sup>10</sup> In this approximation we write the interaction Hamiltonian as<sup>3</sup>

$$\mathcal{H}_I = \sigma_z d \cos \omega t \quad (4)$$

ignoring possible off-diagonal items.<sup>3,12</sup> The quantity  $d$  is just the coupling constant between the field and the TLS. In the physics of low-temperature properties of glasses  $d$  is assumed to be a random quantity, uncorrelated with  $\Delta$  and  $\Lambda$ . This assumption is generally not valid for the case of semiconductors.

In the case of a sound wave  $d = \gamma_{ik} u_{ik}^{(0)}$ , where  $\gamma_{ik}$  is the deformational potential of the TLS and  $u_{ik}^{(0)}$  is the amplitude value of the deformation tensor.

For semiconductors the value of  $\Lambda$  depends on the spatial separation of the wells. In the case of electromagnetic waves  $d = \eta \mathcal{E}_0$ , where  $\eta$  is the dipole moment of the TLS while  $\mathcal{E}_0$  is the amplitude of the electric field.<sup>3,11</sup> For charged particles the dipole moment is proportional to the distance between the wells, and is thereby strongly correlated with the value of  $\Lambda$ .

The total Hamiltonian of the TLS may now be written as<sup>3</sup>

$$\mathcal{H} = (\Delta + d \cos \omega t) \sigma_z - \Lambda \sigma_x. \quad (5)$$

The difference between the eigenvalues of this new Hamiltonian,  $2\epsilon(t)$ , where

$$\epsilon(t) = \sqrt{(\Delta + d \cos \omega t)^2 + \Lambda^2}. \quad (6)$$

is just the energy splitting of the TLS.<sup>13</sup> To characterize a TLS we need not only the energy spacing, but also the occupation numbers of the upper ( $n$ ) and lower ( $1 - n$ ) levels. The non-equilibrium occupation numbers can be found from the balance equation<sup>10</sup>

$$\frac{dn}{dt} = -\frac{n - n_0(t)}{\tau(t)} \quad (7)$$

where  $n_0(t)$  is the adiabatic equilibrium occupation number. It depends on the energy spacing  $\epsilon(t)$ , temperature  $T$  and time  $t$  as

$$n_0(t) = \left[ e^{2\epsilon(t)/k_B T} + 1 \right]^{-1}. \quad (8)$$

The relaxation time  $\tau(t)$  is a function of the energy splitting, the tunneling barrier and temperature, and also depends on the exact relaxation mechanism.

The power absorbed by a single TLS can be determined by the expression<sup>10</sup>

$$p(\Delta, \Lambda) = \frac{2}{\Theta} \int_0^\Theta dt n(t) \frac{d\epsilon}{dt}, \quad \Theta \equiv \frac{2\pi}{\omega}. \quad (9)$$

The contributions of individual TLS must be added and such a summation can be performed in a conventional way using the distribution function  $N(\Delta, \Lambda)$  of the random parameters  $\Delta$  and  $\Lambda$  and replacing the deformational potential  $\gamma_{ik}$  by its average value.

To analyze the nonlinear absorption we use the exact periodic in time solution of Eq. (7) to obtain the following result for the total absorbed power,<sup>10</sup>

$$P = \frac{1}{\Theta} \int_0^\infty \int_0^\infty \frac{N(\Delta, \Lambda) d\Delta d\Lambda}{k_B T} \left( 1 - e^{-\int_0^\Theta dt_1/\tau(t_1)} \right)^{-1} \times \int_0^\Theta \int_0^\Theta \frac{dt dt' \dot{\epsilon}(t) \dot{\epsilon}(t - t')}{\cosh^2[\epsilon(t - t')/k_B T]} e^{-\int_0^{t'} dt_1/\tau(t - t_1)}. \quad (10)$$

Unfortunately, this integral cannot be calculated analytically in the general case. Earlier discussions of the strongly nonlinear regime have considered situations where only very limited time or energy ranges contribute to the absorption. This corresponds to the asymptotic high intensity limits of the integral. Our approach has rather been expanding the integral in powers of the amplitude of the modulation of the energy splitting,  $d$ , to get insights into the first onset of nonlinear effects. So we will concentrate on the regime of weak nonlinearity.

### III. SEMICONDUCTORS IN THE TLS FORMALISM

We will now show how hopping in amorphous or doped crystalline semiconductors can be described by the TLS model.

A TLS is naturally formed by a pair of nearest impurity centers having one electron per pair. To make the calculations for an individual pair we have to specify the coupling constant  $d$  and the relaxation time  $\tau$ . The latter is essentially dependent on the dominant mechanism of pair population relaxation. To sum over all the pairs we have to specify the proper distribution function for the parameters of the pairs containing one electron.

We will concentrate on the case when the interaction has a dipole character and can be expressed in the form

$$d(\mathbf{r}, t) = d_0(\mathbf{r}) \cos \omega t, \quad d_0(\mathbf{r}) \equiv e \mathcal{E}_0 r(\boldsymbol{\nu} \cdot \mathbf{n}) \quad (11)$$

where  $\mathcal{E}_0$  is the amplitude of the effective electric field acting upon the electrons. It is just the amplitude of the local electric field created either by the external electric field, or due to piezoelectric interaction with an acoustic wave.<sup>14</sup>  $\boldsymbol{\nu} = \boldsymbol{\mathcal{E}}/\mathcal{E}$  is the field polarization vector,  $r$  is the distance between the components of the pair,  $\mathbf{n} = \mathbf{r}/r$  is the pair direction vector.

The relaxation time is strongly dependent on the particular mechanism of interaction between localized electrons and phonons, see for example review Ref. 2. Here we will only quote the most important results.

#### A. Relaxation time

In general, the relaxation time can be expressed as<sup>2</sup>

$$\frac{1}{\tau(\epsilon, \mathbf{r})} = \frac{1}{\tau_n(T)} \left( \frac{\epsilon}{k_B T} \right)^n \left( \frac{\Lambda(\mathbf{r})}{\epsilon} \right)^2 \times \Phi_n \left( \frac{2\epsilon}{E_r} \right) \frac{\coth(\epsilon/k_B T)}{[1 + (2\epsilon/E_a)^2]^4}, \quad (12)$$

where the exponent  $n$ , the factor  $\tau_n^{-1}$  and the function  $\Phi_n(x)$  are dependent on the particular mechanism of interaction between localized electrons and phonons. The meaning of the energies  $E_r$  and  $E_a$  will be made clear in a moment.

The energy dependence of  $\tau^{-1}$  can be easily understood. The phonon emitted or absorbed during a transition between the electron levels must obviously have an energy equal to the energy splitting of the pair,  $2\epsilon$ . The power by which it occurs in the formula (12) is determined by the product of the phonon density of states with the frequency dependence of the squared interaction matrix element. The factor  $\coth(\epsilon/k_B T)$  is equal to  $2N_\omega + 1$  where  $N_\omega$  is the Planck function for  $\hbar\omega = 2\epsilon$ . Apart from a proportionality factor, this is the probability of phonon emission,  $\propto N_\omega$ , plus the probability of absorption,  $\propto (N_\omega + 1)$ . The factor  $[\Lambda(\mathbf{r})/\epsilon]^2$  is a dimensionless measure of the tunneling coupling between the bare states in the two wells. It can be seen that  $\tau$  has a minimum with respect to  $\Lambda$  when  $\Lambda = \epsilon$ , which is equivalent to  $\Delta = 0$ . This condition defines the minimal relaxation time  $\tau_0$  referred to in Eq. (2). The minimal  $\tau$  corresponds to the symmetrical configuration when the bare energy levels at both sites are equal. To allow for time dependence of  $\tau$  one should substitute  $\epsilon = \epsilon(t)$  from Eq. (6).

The expression (12) contains two specific energy scales,  $E_a \equiv 2\hbar s/a$ , and  $E_r \equiv \hbar s/r$ . The first scale is the energy of a phonon having a wavelength of the order of the single-site localization length,  $a$ . Phonons with larger energies produce rapidly oscillating fields which average out at the distance occupied by a localized electron. Consequently,  $\tau^{-1}$  strongly decays at  $\epsilon \gtrsim E_a$ . The second scale corresponds to phonons with a wavelength of the order of the distance  $r$  between the components of the pair. If the deformation potentials of both components of the pair are the same, or if the main mechanism of the electron-phonon interaction is piezoelectric, then at  $2\epsilon \ll E_r$  both levels move synchronously, and no interaction occurs. The net interaction is hence proportional to some power of the ratio  $2\epsilon/E_r$ , see Ref. 2 for a review.

In most realistic cases one can assume  $\epsilon \ll E_a$ . However, for  $T \approx 1$  K the ratio  $x = 2\epsilon/E_r$  can be either less or greater than 1, giving different types of behavior for  $\Phi_n(x)$ . We will concentrate on the case of  $x \gg 1$  since this limiting case seems to be more easily accessible for experiments. In this regime,  $\Phi_n(x)$  can be considered as constant. The validity of this approximation will be considered in the Discussion. The quantities  $\tau_n(T)$  are listed in Ref. 2.

The most important feature of the relaxation for our problem is the *energy dependence* of the relaxation rate, namely, the power  $n$ . Under the above-mentioned conditions,  $n = 3$  in the case of deformational interaction and  $n = 1$  for piezoelectric interactions.<sup>2</sup> As will be clear later, only the energies  $\epsilon \ll k_B T$  are important for the anomalous nonlinear behavior, so one can approximate  $\coth(\epsilon/k_B T) \approx k_B T/\epsilon$ . In this way we arrive at the following energy dependences of the relaxation rate:

$$\frac{1}{\tau} \propto \begin{cases} \Lambda^2 \epsilon^0 & \text{for the deformational interaction} \\ \Lambda^2 \epsilon^{-2} & \text{for the piezoelectric interaction} \end{cases} \quad (13)$$

Note that in the first case the relaxation time is indepen-

dent on  $\epsilon$  and thereby on time. This is the same as is the case in dielectric glasses. Apart from a constant and the dependency on the magnetic field, we can thus expect the same type of behavior from these two very different systems. In the second case the  $\epsilon$ -dependence is the same as in metallic glasses, which has also been analyzed by the authors<sup>9</sup> and has proven to be the source of a pronounced anomalous effect. In particular, in metallic glasses the lowest nonlinear contribution is proportional to the intensity to 3/2 rather than to the intensity squared as for dielectric glasses.

In the absence of the magnetic field the energy overlap integral  $\Lambda$  is related to the distance  $r$  between the sites of a pair simply as  $\Lambda = \Lambda_0 \exp^{-r/a}$ , where  $\Lambda_0 = (1 - 5) \times m e^4 / \hbar^2 \kappa^2$  is of the order of the effective Bohr energy. Here  $m$  is the electron effective mass while  $\kappa$  is the dielectric constant. A magnetic field will squeeze the electron wave function, and this effect will be strongest for the direction perpendicular to the field. This introduces an angular dependency to the localization length and thereby also to  $\Lambda$ . Following Ref. 15, we will analyze the limiting cases of weak (w) and strong (s) magnetic field where the influence of magnetic field is weak or strong, respectively. The asymptotic expressions for the  $\zeta(\mathbf{r}) \equiv -\ln[\Lambda(\mathbf{r})/\Lambda_0]$  are the following<sup>15</sup>:

$$\zeta_w = \frac{r}{a} + \frac{r^3 a \sin^2 \theta}{24 \lambda^4}, \quad (14)$$

$$\zeta_s = \frac{r^2 \sin^2 \theta}{4 \lambda^2} + \frac{|r \cos \theta|}{a_H}. \quad (15)$$

Here  $\theta$  is the angle between  $\mathbf{r}$  and the direction of the magnetic field  $\mathbf{H}$ ,  $\lambda = \sqrt{\hbar c / e H}$  is the magnetic length, while  $a_H = \hbar / \sqrt{2m E_H}$  is the characteristic localization length in the longitudinal direction, where  $E_H$  is the ionization energy of the ground state of the localized electron in the magnetic field.

## B. Pair distribution function

The total absorption is given by a sum of the contributions of the individual pairs. Hence we have to sum over the  $\mathbf{r}$ , as well as over the individual energies of the electron levels. The latter summation must take into account the correlation between the level occupation numbers due to Coulomb interaction. As shown in Ref. 16, the summation over the energies can be split into integration over the pair center-of-gravity and over the bare inter-level spacing  $\Delta$ . The first integration gives  $2\Delta + e^2/\kappa r$  since only the pairs with the center-of-gravity energy between the chemical potential  $\mu + \Delta$  and  $\mu - \Delta - e^2/\kappa r$  have one electron per pair. As a result, the pair distribution function can be expressed through the single-electron density of states  $g$  as<sup>16</sup>,

$$N(\Delta, \mathbf{r}) = \frac{g^2 V}{4\pi} \left( 2\Delta + \frac{e^2}{\kappa r} \right) \quad (16)$$

where  $V$  is the volume contributing to the absorption. This expression is valid if for a typical hopping distance  $e^2/\kappa r \gg \Delta_C$  where  $\Delta_C$  is the width of the *Coulomb gap* in the single-electron density of states.<sup>16</sup> Inside the Coulomb gap, we would rather have to use the distribution

$$N(\Delta, \mathbf{r}) = \frac{3}{40\pi^3} \left(\frac{\kappa}{e^2}\right)^6 \left(2\Delta + \frac{e^2}{\kappa r}\right)^5. \quad (17)$$

Calculations for both cases are similar. Note that the distribution function is isotropic. Under the conditions of interest to us the typical hopping distance  $r$  is small enough to let us neglect  $2\Delta$  in comparison with  $e^2/\kappa r$  in Eqs. (16) and (17). Thus the distribution becomes  $\Delta$ -independent, and we denote it as  $N(\mathbf{r})$ . Using the no-

tation presented above we can now write the coupling constants  $\alpha_1, \alpha_2 \approx \alpha$  from Eqs. 1, and 2 as<sup>17</sup>

$$\alpha = \frac{4\pi^3}{3} \mathcal{K}^2 \frac{e^4 g^2 a r_\omega^3}{\kappa^2}, \quad r_\omega = a \ln \frac{\Lambda_0}{\hbar\omega}, \quad (18)$$

where  $\mathcal{K}$  is the coupling constant of the piezoelectric interaction. The power of  $r_\omega$  may vary for different type of interactions, depending on whether the interaction includes the dipole moment of the pair.

#### IV. CALCULATION OF ABSORPTION

As a result of the previous considerations, the absorbed power can be expressed as

$$P = \int d\mathbf{n} \int_0^\infty d\Delta \int_0^\infty r^2 dr N(r) \left\{ \int_0^\Theta \int_0^\Theta \frac{dt dt'}{\Theta k_B T} \frac{\dot{\epsilon}(\mathbf{r}, t) \dot{\epsilon}(\mathbf{r}, t-t')}{\cosh^2[\epsilon(\mathbf{r}, t-t')/k_B T]} \frac{\exp\left(-\int_0^{t'} dt_1/\tau(t-t_1)\right)}{1 - \exp[-\int_0^\Theta dt_1/\tau(t_1)]} \right\}. \quad (19)$$

Here the expression in the braces is just the power absorbed by an individual pair,  $p(\Delta, \mathbf{r})$ . The energy splitting  $2\epsilon$  depends on  $\mathbf{r}$  through the interaction potential  $d_0(\mathbf{r})$  given by Eq. (11) and through the tunneling splitting  $\Lambda = \Lambda_0 e^{-\zeta(\mathbf{r})}$ . Technically it is convenient to transform the integral from the set of variables  $r, \mathbf{n}$ , to the variables  $\Lambda, \theta, \phi$ . Such a transform introduces the factor

$$\left| \frac{\partial \Lambda(r, \mathbf{n})}{\partial r} \right|^{-1} = \frac{a}{\Lambda} f(\Lambda, \mathbf{n}).$$

Here the dimensionless function  $f(\Lambda, \mathbf{n})$  is given by the equation

$$f(\Lambda, \mathbf{n}) \equiv -\frac{1}{a} \frac{\partial r_\Lambda(\mathbf{n})}{\partial \ln \Lambda} = \frac{1}{a} \frac{\partial r_\Lambda(\mathbf{n})}{\partial \mathcal{L}}. \quad (20)$$

where  $\mathcal{L} \equiv \ln(\Lambda_0/\Lambda)$ , while  $r_\Lambda(\mathbf{n})$  is the solution of the equation

$$\zeta(r_\Lambda, \mathbf{n}) = \mathcal{L}. \quad (21)$$

In the simplest case of zero magnetic field  $f = 1$ , and we have a distribution very similar to those of glasses. The variable transform strongly simplifies the calculations since the relaxation time is a function of the parameters  $\Delta$  and  $\Lambda$  and the quantity  $r_\Lambda$  is a weak (logarithmic) function of  $\Lambda$ . Consequently it can be extracted out of the integral over  $\Lambda$ , while replacing  $\Lambda$  in the expression for  $r_\Lambda$  by its characteristic value. As a result, the integrations over  $\Delta$  and  $\Lambda$  will remain the same as previously calculated for glasses, and only the angular integration and the dependence on the characteristic value of  $\Lambda$  are different.

The following calculation procedure is similar to that of Ref. 17. The expression (19) will be expanded in powers of the effective electric field, and the lowest correction will be compared with the linear result. Expanding the individual contributions in powers of  $d_0$  as

$$p(\Delta, \mathbf{r}) = \sum_{k=2}^4 p^{(k)} d_0^k(\mathbf{r}) \quad (22)$$

we notice the coefficients  $p^{(k)}$  depend only on the quantities  $\Delta$  and  $\Lambda$ . Transforming the variables from  $r, \mathbf{n}$  to  $\Lambda, \mathbf{n}$  we can use the fact that  $r$  is a weak function of  $\Lambda$  and extract of the quantities proportional to the powers of  $r$  from the integral over  $\Lambda$  replacing

$$r \rightarrow r_c(\mathbf{n}) = r_\Lambda(\mathbf{n})|_{\Lambda=\Lambda_c}$$

where  $\Lambda_c$  is the characteristic value determined by the integrand over  $\Lambda$ . In a similar way, we replace  $f(\Lambda, \mathbf{n}) \rightarrow f_c(\mathbf{n}) = f(\Lambda_c, \mathbf{n})$ . Finally we arrive at the expression  $P = \sum_{k=2}^4 P^{(k)}$  with  $P^{(k)} = (ae\mathcal{E}_0)^k I_k J_k$  where

$$I_k = \int_0^\infty d\Delta \int_0^\infty d\Lambda \Lambda^{-1} p^{(k)}(\Delta, \Lambda), \quad (23)$$

$$J_k = \int d\mathbf{n} (\boldsymbol{\nu} \cdot \mathbf{n})^k f_c(\mathbf{n}) N(r_c) (r_c/a)^k \quad (24)$$

The quantities  $I_k$  are the same that enter the expressions for nonlinear absorption in glasses, and we quote them from Ref. 9.

Parallel with  $I_k$  we can also estimate  $\Lambda_c$ , and thereby  $\mathcal{L}$ . It can be shown that this value is only weakly dependent on the mechanism of absorption and on  $k$ . For all the mechanisms analyzed in this paper we have

$$\mathcal{L} = \ln \frac{c_{\mathcal{L}} \Lambda_0}{k_B T \sqrt{\omega \tau_n(T)}}. \quad (25)$$

The value for  $c_{\mathcal{L}}$  is of the order one, and varies only with a factor less than two for the situations discussed in this paper. We will therefore treat  $\mathcal{L}$  as independent of  $k$  for this discussion.

In this paper we will present the results for the case of low frequencies,  $\omega \tau_n \ll 1$ , for which the anomalous nonlinear behavior is most pronounced. In this case the linear in the intensity contribution,  $I_0$ , is independent of  $\tau$ , and moreover, it is the same for any dependence  $\tau(\epsilon)$ ,

$$I_0 = (\pi^2/16) \omega \approx 0.62 \omega. \quad (26)$$

The case of deformational interaction, for which the parameter  $n$  in Eq. (12) is equal to 3, corresponds to the situation in dielectric glasses. In this case<sup>9</sup>

$$I_3 = 0, \quad I_4 \approx 0.054 \frac{\omega}{\sqrt{\omega \tau_3(T)} (k_B T)^2}. \quad (27)$$

The piezoelectric interaction ( $n = 1$ ) is similar to the case of metallic glasses. It can be shown that the energy dependence of the relaxation time leads to a divergence in the integration over  $\Lambda$  and  $\Delta$ . To get a proper estimate one should cut off the integration at  $\epsilon \lesssim d_0$ . As a result, the leading nonlinear contribution appears proportional to  $|d_0|^3$  and equal to<sup>9</sup>

$$I_3 \approx 0.1 \omega / k_B T. \quad (28)$$

We again expect  $\mathcal{L}$  to be given by Eq. 25. To calculate the angular integrals  $J_k$  we solve Eq. (21) in a recursive way. For the case of weak magnetic fields, we obtain

$$r_c = a \mathcal{L} (1 - \mathcal{L}^2 a^4 \sin^2 \theta / 24 \lambda^4); \quad (29)$$

$$f_c = 1 + a^4 \mathcal{L}^2 \cos^2 \theta / 8 \lambda^4. \quad (30)$$

For the strong field limit, solving a quadratic equation, we obtain

$$r_c = \frac{2 \lambda^2 \cos \theta}{a_H \sin^2 \theta} \left( \sqrt{1 + \tan^2 \theta \mathcal{L} (a_H / \lambda)^2} - 1 \right); \quad (31)$$

$$f_c = (a_H / a) \mathcal{L} \cos \theta (1 + \mathcal{L} (a_H / \lambda)^2 \tan^2 \theta)^{-1/2}. \quad (32)$$

Here  $\theta$  is the angle between  $\mathbf{n}$  and the direction of magnetic field  $\mathbf{H}$ . Hence,  $d\mathbf{n} = d(\cos \theta) d\phi$  where  $\phi$  is the azimuthal angle between the projections of  $\mathbf{n}$  and  $\mathbf{E}_0$  on the plane, perpendicular to  $\mathbf{H}$ . After substitution of Eqs. (29)–(32) into Eq. (24) the angular integrals are calculated directly.

## V. RESULTS

To set the scale of nonlinear corrections let us start with the expression for the linear absorption in the absence of magnetic field,  $P_0 \equiv P_0(0)$ . Both linear and nonlinear contributions are dependent on the relaxation mechanism of the relevant pairs.

### A. Deformational interaction between localized pairs and phonons

We will first consider deformational interaction between the localized pairs and thermal phonons. For the deformational mechanism,

$$P_0 = (\pi^2/48) (V a^4 g^2 e^4 \omega \mathcal{L}^3 \mathcal{E}_0^2 / \kappa). \quad (33)$$

In the absence of magnetic field we obtain

$$P_4(0) = \frac{P(0) - P_0(0)}{P_0(0)} = 0.26 \frac{F^2 \mathcal{L}_c^2}{\sqrt{\omega \tau_0(T)}} \quad (34)$$

were we have introduced the dimensionless “field amplitude”

$$F \equiv e \mathcal{E}_0 a / k_B T. \quad (35)$$

In a weak magnetic field, the quadratic in magnetic field corrections arise both to the linear absorption and to the lowest nonlinear contribution. They can be expressed in a unified way as, cf. with Ref. 17,

$$P_{0/4}^{(w)}(H) = P_{0/4}(0) [1 - c_w (a/\lambda)^4 \mathcal{L}^2], \quad (36)$$

so the magnetic field produces corrections which are  $\propto H^2$ . The numerical factor  $c_w$  depends on the direction of the electric field  $\mathbf{E}_0$  with respect to the magnetic field  $\mathbf{H}$ . Its values are also different for the linear and nonlinear, contributions,  $c_w^{(0)}$  and  $c_w^{(4)}$ , respectively. The values of  $c_w$  are shown in Table I.

Direction	$c_w^{(0)}$	$c_w^{(4)}$	$c_w^{(3)}$
$\mathbf{H} \perp \mathbf{E}_0$	0.2	$\approx 0.29$	$\approx 0.25$
$\mathbf{H} \parallel \mathbf{E}_0$	0.1	$\approx 0.095$	$\approx 0.097$

Table I. Numerical coefficients entering the nonlinear contributions to the absorption.

The decrease of attenuation in the magnetic field has the following physical reason. The presence of a magnetic field squeezes the electron wave functions, and the overlap integrals between the components of the pair decrease. Furthermore, the wave functions are squeezed mostly in the direction perpendicular to  $\mathbf{H}$ . On the other hand, the coupling between the wave and the pair is maximal if the pair dipole moment is parallel to  $\mathbf{E}_0$ . Thus the reduction of absorption is more pronounced for  $\mathbf{H} \perp \mathbf{n}$ . For the nonlinear contribution the difference should be even stronger, as it includes higher orders of the dipole moment.

In the limit of strong magnetic fields the results are even more interesting, as the functional dependency on

the magnetic field also varies with the different absorption types. Yet we still see the same relative considerations as for the weak field limit. For a magnetic field parallel to the radiation polarization vector we get

$$\frac{P_0^{(s)}(H)}{P_0} = 3 \frac{\lambda^2 a_H^2}{\mathcal{L} a^4} \propto H^{-4/3}, \quad (37)$$

$$\frac{P_4^{(s)}(H)}{P_4(0)} = \frac{5}{2} \frac{\lambda^2 a_H^4}{\mathcal{L} a^6} \propto H^{-5/3}. \quad (38)$$

For a perpendicular field the results show both a stronger dependency on  $H$  and on the order of the expansion in intensity.

$$\frac{P_0^{(s)}(H)}{P_0(0)} = 6 \frac{\lambda^4}{\mathcal{L}^2 a^4} \ln \frac{\mathcal{L} a_H^2}{\lambda^2} \propto H^{-2} \ln H, \quad (39)$$

$$\frac{P_4^{(s)}(H)}{P_4(0)} = 30 \frac{\lambda^6}{\mathcal{L}^3 a^6} \ln \frac{\mathcal{L} a_H^2}{\lambda^2} \propto H^{-3} \ln H \quad (40)$$

### B. Piezoelectric interaction between localized pairs and phonons

We will now turn our attention to the piezoelectric interaction. This also interacts via a dipole moment, so the linear results are basically the same, apart from a coupling constant. However, there is a striking difference for the nonlinear contribution – similarly to the case of metallic glasses,<sup>9</sup> the integration over  $\Delta$  and  $\Lambda$  in Eq. (23) results in a term proportional to  $|d_0|^3$  rather than  $d_0^4$ . This is reflected in a change of the absorption dependencies both on the wave intensity and on the magnetic field. For this case, we restrict ourselves by order-of-magnitude estimates for numerical factors. Calculation of exact numbers would require a great amount of numerical work which would be inadequate to the accuracy of the initial model for the electron density of states. The results read as,

$$P_3(0) = [P - P(0)]/P_0 = c_m |F|, \quad (41)$$

where the estimate for  $c_m$  is 0.1. Note that in this case the expansion of the absorption in powers of intensity appears *non-analytical* which implies relatively strong non-linearity.

Since now the expansion in  $d$  of the nonlinear contribution starts with  $|d_0|^3$  rather than from  $d_0^4$  one can expect weaker magnetic field effects. This is indeed the case. The values of the numerical coefficients  $c_w^{(3)}$  are shown in the Table I.

For strong fields we get for parallel and perpendicular fields respectively

$$\left( \frac{P_3^{(s)}(H)}{P_3(0)} \right)_{\parallel} = \frac{8}{3} \frac{\lambda^2 a_H^3}{\mathcal{L} a^5} \propto H^{-3/2} \quad (42)$$

$$\left( \frac{P_3^{(s)}(H)}{P_3(0)} \right)_{\perp} = \frac{64}{3} \frac{\lambda^5}{\mathcal{L}^{5/2} a^5} \ln \frac{\mathcal{L} a_H^2}{\lambda^2} \propto H^{-5/2} \ln H. \quad (43)$$

Let us recall that the above expressions are valid for the low frequency limit only, where  $\omega\tau_n(T) \ll 1$ . Similar calculations are possible for the high frequency limit, as well as for different pair distribution functions. In particular, for the case of pronounced Coulomb gap the essential differences occur only in powers of  $r$  and thereby of  $\mathcal{L}$ . Thus the influence of the magnetic field will be different. The relation between linear and nonlinear results remain similar, apart from numerical factors.

## VI. DISCUSSION

Let us discuss the relevance of the obtained results for realistic materials and situations. In this connection, several parameters are to be considered.

Regarding the material properties, we consider only *weakly doped* or amorphous semiconductors in the regime of *nearest-neighbor hopping conductance*. Hence, we calculate absorption by close pairs independent of each other. To keep the model adequate we have to require that the typical inter-center distance within the pair,  $r_c$ , should be much smaller than the typical distance between defect centers,  $\bar{r} = (4\pi n_d/3)^{-1/3}$ . Here  $n_d$  is the defect concentration. The *hopping distance*  $r_c$  is discussed in Sec. IV.

Another requirement is that the impurities are not too shallow, so that the electrons cannot be excited from the localized states to the conduction band by the AC perturbing potential. There are experimental examples of this, where such an excitation serves as a source of nonlinear behavior.<sup>6</sup>

According to the present calculation, the most interesting effects occur at “low” frequencies when  $\omega\tau_n(T) \ll 1$ . This requirement also ensures that the relaxation absorption dominates the resonant one. Certainly, the minimal relaxation time  $\tau_n(T)$  is a material property. Usually the above inequality is met at low temperatures for frequencies in the range 100 – 1000 MHz.

The TLS model in glasses is restricted to very low temperatures where higher energy levels are not excited. The situation is a bit different in semiconductor materials where the inter-level splittings are of the order of the Bohr energy. Consequently, the nearest-neighbor hopping conductance can be effective in the temperature range up to a few K.

The main objectives of this paper is to show that nonlinear effects are anomalously large. Indeed, in the case of deformational absorption an additional parameter  $(\omega\tau_3)^{-1/2} \gg 1$  is present in the nonlinear expansion (34), while the the case of piezoelectric interaction the nonlinear expansion starts from the *first power* of dimensionless amplitude  $|F|$ , Eq. (41). Furthermore, the nonlinear contributions have pronounced magnetic field dependences

different from the linear ones. We hope that those features will allow experimentalists to detect the nonlinear behavior and to discriminate between different relaxation mechanisms for localized states.

In course of the present calculations we have assumed the inequality  $\epsilon/E_r \gg 1$  to be met, see Eq.(12). It is important that for  $\epsilon$  one has to substitute the value which gives the dominating contribution in the final integration over the pair distribution function. This value is actually different for the linear and the nonlinear contributions, and it is also dependent on the relaxation mechanism. It turns out that for the linear absorption this typical  $\epsilon \sim k_B T$ , while for the nonlinear contributions it is reduced by a factor  $\sqrt{\omega\tau_3}$  for the deformational interaction, by  $|F|$  for the piezoelectric one, both calculated under the condition  $\epsilon \gg E_r$ . A similar estimate is necessary to choose a proper value for  $r$  in the expression for  $E_r$ . This value depends on the quantities  $\sqrt{\omega\tau_n}$ ,  $\Lambda_0$ , as well as on the magnetic field. Thus the experimental variables intensity, frequency and magnetic field, in addition to the system parameters  $\tau_0$  and  $\Lambda_0$ , influence the behavior of the nonlinear absorption. This rich parameter space allows for a large range of experiments.

## VII. CONCLUSIONS

In this paper, we have analyzed nonlinear contributions to the acoustic and electromagnetic absorption by localized electron states in semiconductors in the regime of hopping conductance. The most important conclusions are the following.

- The total behavior of absorption is determined by the product  $\omega\tau$  where  $\tau(T)$  is the minimal relaxation time for a pair with energy splitting of the order  $k_B T$ .
- The anomalous nonlinear behavior occurs at  $\omega\tau \ll 1$ . In the case of deformational relaxation mechanism for the localized electrons a large additional factor  $(\omega\tau)^{-1/2}$  appears in front of the item  $\propto F^2$  in the expansion of nonlinear absorption. In the case of piezoelectric relaxation mechanism the expansion starts with the item  $\propto |F|$  rather than  $\propto F^2$ . Here  $F$  is the dimensional AC field amplitude.
- The anomalous nonlinear absorption is strongly influenced by an external magnetic field, the influence being dependent both on the electron pair distribution function, on the dominating relaxation mechanism for the localized electrons, and on the direction of the field polarization vector with respect to the magnetic field. The influence of magnetic field on the linear absorption and nonlinear corrections is substantially different.

As a result, the physical picture of weakly nonlinear absorption appears rich and informative. Our estimates

show that the effects under consideration are accessible for the modern experiment, and many important characteristics – the dominating relaxation mechanism, the importance of the Coulomb gap, typical hopping distances, etc. – can be extracted by comparison to the present theory provided the experiment will be done.

It should be emphasized that there is a close similarity between the present and the results of our previous calculations for glassy materials.<sup>9</sup> However, the localized states in disordered semiconductors, being charged, can be influenced by magnetic field which makes them easier to investigate. We therefore also hope that the studies of semiconductor systems will also provide a new information regarding nonlinear response of TLSs in glasses.

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